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Letter

Tunneling Properties at the Interface between Superconducting Sr_2RuO_4 and a Ru Microinclusion

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We have investigated the magnetic field and temperature dependence of the tunneling spectra of the eutectic system Sr_2RuO_4-Ru. Electric contacts to individual Ru lamellae embedded in Sr_2RuO_4 enable the tunneling spectra at the interface between ruthenate and a Ru microinclusion to be measured. A zero bias conductance peak (ZBCP) was observed in the bias voltage dependence of the differential conductance, suggesting that Andreev bound states are present at the interface. The ZBCP starts to appear at a temperature well below the superconducting transition temperature. The onset magnetic field of the ZBCP is also considerably smaller than the upper critical field when the magnetic field is parallel to the *ab*-plane. We propose that the difference between the onset of the ZBCP and the onset of superconductivity can be understood in terms of the existence of the single-component state predicted by Sigrist and Monien.

KEYWORDS: Sr_2RuO_4, p-wave superconductor, eutectic, zero bias conductance peak

Recent experimental and theoretical studies have revealed that Sr_2RuO_4 is a spin-triplet superconductor.[1,2] An NMR measurement confirmed this first: the Knight shift of ^{17}O is not affected by the superconducting (SC) transition, indicating that the spin state of the Cooper pair is triplet.[3] It was demonstrated by a muon spin relaxation measurement[4] that spontaneous magnetic moments accompany the SC transition. This result indicates that time reversal symmetry is broken in the SC phase, suggesting that the orbital part of the order parameter of the SC state has two components with a relative phase of $\pi/2$: $k_x + ik_y$.

Amongst the many interesting SC properties of Sr_2RuO_4, the enhancement of the SC transition temperature in the eutectic system[5] is rather interesting. Under particular growth conditions, a single crystal of Sr_2RuO_4 with Ru lamellae included is obtained. This eutectic system, Sr_2RuO_4-Ru, shows a broad SC transition with an onset of approximately 3 K,[5] called the 3-K phase. The manifestation of 3-K phase superconductivity has been confirmed by ac susceptibility measurements[5] and resistivity measurements.[5,7] These experimental studies suggest that 3-K phase superconductivity is inhomogeneous and filamentary, and thus it is inferred to occur at the interface between Sr_2RuO_4 and Ru.

The anisotropy of the upper critical field indicates that the Sr_2RuO_4 side of the interface essentially sustains the superconductivity.[5] The temperature dependence of the upper critical magnetic field H_{c2} shows an unusual upturn at low temperatures when the magnetic field is applied parallel to the c-axis.[8] The low-temperature enhancement of H_{c2} for $H \parallel c$ can be explained in terms of the coupling of the two components of the order parameter discussed in Sigrist and Monien's theory.[9] The good agreement between theory and experiment supports the description of the 3-K phase by a two-component order parameter with a relative phase of $\pi/2$. According to the theory by Sigrist and Monien, a single-component order parameter k_x with its lobes parallel to the interface shown in Fig. 1(a), nucleates at the onset of the 3-K phase. With decreasing temperature and/or magnetic field, the superconductor undergoes another transition to the two-component state $k_x + i\varepsilon k_y$ ($0 < \varepsilon < 1$) with broken time reversal symmetry, similar to pure Sr_2RuO_4 without Ru inclusions.

Tunneling spectroscopy has played a crucial role in determining the phase structure of d-wave superconductors,[10–12] and thus will be a promising technique for examination of the transition to the two-component state in the 3-K phase. Several tunneling spectroscopy measurements have already been performed on Sr_2RuO_4.[13,14] A tunneling measurement using cleaved junctions of the eutectic system Sr_2RuO_4-Ru was demonstrated by Mao *et al.*[15] Their results gave strong evidence that unconventional superconductivity occurs in the 3-K phase. They observed a zero bias conductance peak (ZBCP) in the voltage dependence of the differential conductance. The ZBCP is interpreted in terms of an Andreev bound state (ABS) at the interface between Sr_2RuO_4 and Ru.

In this paper, our experimental study of the ZBCP in the 3-K phase is reported. A microfabrication technique enabled the measurement of the differential conductance at the interface between a single Ru microinclusion and Sr_2RuO_4. The ZBCP was observed in the voltage dependence of the differential conductance. We have found that the ZBCP starts to appear at a lower temperature and/or magnetic field than the onset of 3-K phase superconductivity. The difference between the onset of the ZBCP and the onset of 3-K phase superconductivity is discussed with the help of Sigrist and Monien's theory.[9]

The eutectic samples of Sr_2RuO_4-Ru used in the present experiment were grown by a floating zone

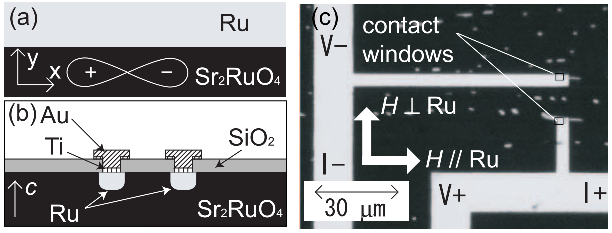

Fig. 1. (a) Schematic of the interface between Sr_2RuO_4 and Ru modeled by Sigrist and Monien. (b) Cross-sectional sketch of the sample. Sample structure is described in detail in text. (c) Top view of the sample used. The c-axis direction is normal to the picture plane. The contact windows are seen at the edge of Ti/Au electrodes. Magnetic field directions with respect to the Ru lamella are indicated by arrows.

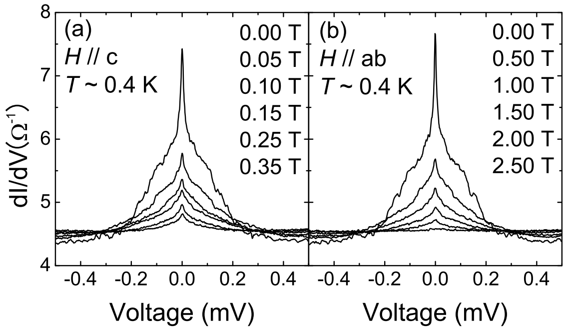

Fig. 2. ZBCP observed in the bias voltage dependence of $\mathrm{d}I/\mathrm{d}V$ in sample A at approximately 0.4 K. Magnetic field direction is parallel to the c-axis in (a) and parallel to the ab-plane and the Ru-plane in (b).

method. Ru lamellae with typical dimensions of approximately 1 μm × 5 μm × 20 μm were included in the samples. Details of the crystal growth and the properties of the eutectic samples are described in Ref. 5. In order to measure the conductance at the interface between a single Ru microinclusion and Sr_2RuO_4, we fabricated the device illustrated in Fig. 1(b). The device has pairs of Ti/Au electrodes directly attached to individual Ru microinclusions through 2 μm × 3 μm contact windows. Figure 1(c) is a top view of the device. This device was fabricated by employing the following microfabrication technique. First, a 200-nm-thick SiO_2 insulating film was deposited on the polished ab-plane surface of the Sr_2RuO_4-Ru eutectic by rf sputtering. Contact windows of 2 μm × 3 μm were etched through the SiO_2 film by using standard electron beam lithography and an Ar ion milling technique, such that the contact windows were placed immediately above the Ru lamellae. The positions of the Ru lamellae were accurately measured in advance by an optical microscope which has a sample stage equipped with a laser interferometer. Finally, electrodes of Ti/Au were patterned by means of electron beam lithography and a lift-off technique. The thicknesses of the Ti and Au films were about 20 nm and 80 nm, respectively.

As seen in Fig. 1(c), the contact windows are slightly larger than the Ru lamellae. However, a non-superconducting surface layer with a relatively high resistivity exists at the polished surface of the Sr_2RuO_4 crystal. It is reported that the resistance between Sr_2RuO_4 and an In wire with a cross-sectional area of 0.05 mm^2 ranges from 0.1 to 100 Ω.[17] This leads to the estimated resistance through the (2-μm × 3-μm)-sized contact window to be between 1 kΩ and 1 MΩ. The contact resistance between the electrodes and the Ru metal lamella is expected to be much smaller than that between the electrodes and the non-superconducting surface layer. Therefore, the measurement current will dominantly flow through the Ru metal lamella.

The differential conductance between two Ru lamellae was measured by a two-terminal method as in Fig.1(c). A lock-in technique was employed with alternating currents of 10 μA at 184 Hz. Temperature was decreased down to 0.3 K by means of a ^{3}He refrigerator. A magnetic field of up to 4 T was applied by means of a superconducting solenoid. A single-axis sample rotator enabled the magnetic field to be aligned parallel to the c-axis or the ab-plane, within a few degrees.

We measured the tunneling spectra on three junction samples with different conductances on the same crystal. The conductances of the three samples A, B and C were 7.5, 36 and 0.28 Ω^{-1}, respectively. Tunneling spectra with a sharp conductance peak at zero bias voltage were obtained for sample A as shown in Fig. 2; the bias voltage dependence of the differential conductance $\mathrm{d}I/\mathrm{d}V$ at approximately 0.4 K is plotted for different magnetic fields. The magnetic field direction is parallel to the c-axis in (a) and parallel to the ab-plane and the Ru lamella plane in (b). The field direction with respect to the Ru lamella plane is illustrated in Fig. 1(c). The ZBCP persists to well above the upper critical field of the 1.5-K phase and thus involves the 3-K phase.

Due to the Joule heating by the bias voltage, the temperature increased during the measurement. The increase of temperature was no greater than 0.1 K at 0.5 mV. The effect of the Joule heating was negligible at the voltages below 0.1 mV. Because the width of the ZBCP is smaller than 0.2 mV, we believe that the Joule heating affected the feature of the ZBCP negligibly.

The presence of the ZBCP in the spectra ensures that an ABS is formed at the interface involving the Ru lamellae. In our measurement configuration, the resistances of Sr_2RuO_4, Ru and Ti/Au electrodes are measured in series. The resistance of Sr_2RuO_4 estimated from its resistivity $\rho_{ab} \approx 1$ $\mu\Omega$cm and $\rho_c \approx 30$ $\mu\Omega$cm is much smaller than the measured resistance. The measured resistance is probably dominated by the resistance at the Sr_2RuO_4/Ru interface. ZBCP is absent in samples B and C. Although the magnetic field dependence of the conductance shows a small peak in sample B, it can be suppressed by the application of a much smaller field (0.2 T) than H_{c2} of the 3-K phase. Therefore, the peak is considered to have an origin other than the 3-K phase. Sample C shows a conductance dip rather than a peak in the voltage dependence of the differential conductance. This feature is characteristic of the Andreev reflection at an interface with a large tunnel barrier. This suggests that the condition of the interface may differ from place to

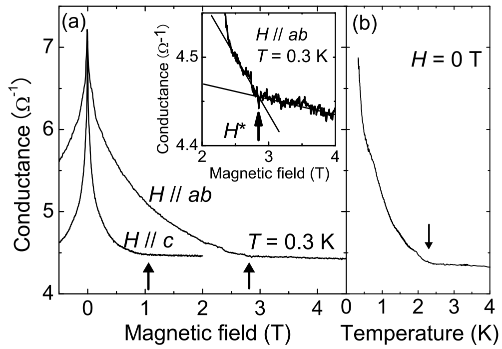

Fig. 3. (a) Magnetic field dependence of the zero bias conductance at $T = 0.3$ K for $H \parallel c$ (lower curve) and $H \parallel ab \parallel$ Ru-plane (upper curve). The onset of the ZBCP is indicated by arrows. The inset shows the definition of the onset magnetic field of the ZBCP H^*. (b) Temperature dependence of the zero bias conductance in zero magnetic field. The onset of the ZBCP is indicated by an arrow.

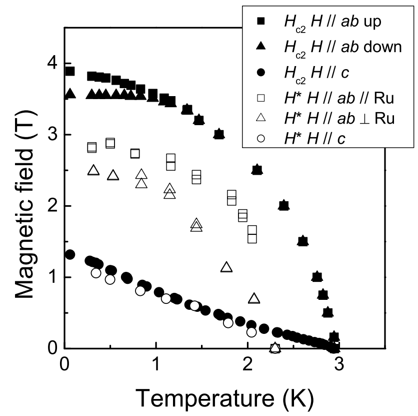

Fig. 4. Temperature dependence of the onset of the ZBCP H^* and the upper critical magnetic field H_{c2}. Closed symbols represent H_{c2} and open symbols represent H^*.

place within a single piece of crystal. Further systematic experimental studies are awaited to allow the condition for the ZBCP to be identified.

Tunneling spectra on sample A are similar to those in the cleaved junctions in Ref. 15. Sharp conductance peaks together with bell-shaped broad conductance peaks are seen in zero magnetic field in both experiments. However, there are still quantitative differences between the results reported in Ref. 15 and those for the present experiment. The width of the conductance peak in our experiment is about half that reported in Ref. 15. The quantitative difference between the two experiments might be attributed to the difference in the sample structure. In our sample, the conductance associated with two Sr_2RuO_4/Ru interfaces in series was measured. In the cleaved junctions in Ref. 15, many Sr_2RuO_4/Ru interfaces at the cleavage were probably measured in parallel. The size and the number of junctions involved cannot be specified in the cleaved junction experiment, making it difficult to compare the two experiments quantitatively.

The magnetic field dependence of the ZBCP shows distinctly different behavior for different field directions. Figure 3(a) shows the magnetic field dependence of the conductance at $T = 0.3$ K for $H \parallel c$ and $H \parallel ab \parallel$ Ru-plane. Here we pay attention to the magnetic field H^* at which the conductance starts to increase with decreasing magnetic field. H^* is defined as the intersection of two tangential lines as shown in the inset of Fig. 3(a). It should be noted that H^* corresponds to the onset magnetic field of the ZBCP, which is consistent with the spectra in Fig. 2. We compare the temperature dependence of H^* with that of H_{c2}. The temperature dependence of H^* is plotted in Fig. 4 with open symbols together with the temperature dependence of H_{c2}. The data for H_{c2} is taken from Ref.16. Similar to the definition of H^*, H_{c2} in Ref.16 is defined as the onset of the resistance drop in a bulk sample from a different batch. On the other hand, H_{c2} in Ref.8 is defined as the inflection point associated with the resistance drop. There are remarkable differences between H^* and H_{c2}. The temperature dependence of the upper critical field H_{c2} shows hysteresis at low temperatures only when magnetic field is applied parallel to the ab-plane.[7,8] However, no clear hysteresis can be seen in the temperature dependence of H^*. Despite the difference of the sample batch, $H^*(T)$ and $H_{c2}(T)$ show a good coincidence for $H \parallel c$ except at very low fields. On the other hand, $H^*(T)$ is considerably smaller than $H_{c2}(T)$ for $H \parallel ab$ and $H \approx 0$ T. We suggest that the ZBCPs are absent near the onset of the 3-K phase in a magnetic field $H \parallel ab$ and $H \approx 0$. While the onset temperature of the ZBCP in the zero magnetic field is 2.3 K as shown in Fig. 3(b), a standard resistance measurement on a sample from the same batch has yielded $T_c = 2.7$ K (onset), higher than the onset of the ZBCP. This also supports the above conclusion.

Superconducting properties of the 3-K phase such as T_c and H_{c2} may differ from place to place, which would cause a discrepancy between H^* and H_{c2}. However, there are reasons why H^* and H_{c2} are different in origin, as suggested in Fig.4. The phase diagrams in Refs.7 and 8 are very similar and H_{c2} exhibits hysteresis for $H \parallel ab$, unlike H^*. In fact, it would be desirable to determine both H_{c2} and H^* in the same junction. However, the resistance measured was dominated by the interface resistance, so that the resistance change due to the SC transition was barely detected.

We propose that the difference between H^* and H_{c2} can be attributed to the phase-sensitive nature of the ABS.[11,12] When Andreev reflection occurs at a superconductor/normal-metal interface, the reflected quasiparticle gains the phase of the pair potential. Because an ABS is a consequence of the interference of the injected and reflected quasiparticles, the condition for the formation of the ABS strongly depends on the phase structure of the pair potential. According to a theory for a normal metal/anisotropic superconductor junction,[11] the ABS is not formed for any incident angle in the case of a single-component order parameter k_x with its lobes parallel to the interface, as depicted in Fig. 1(a). On the other hand, the other component k_y with its lobes perpendicular to the interface gives rise to the ZBCP. The ZBCP is also theoretically predicted in the case of the chiral order parameter $k_x + i\varepsilon k_y$.[18,19] Thus the appear-

ance of the ZBCP is dependent on the phase structure of the pair potential. The absence of the ZBCP near the onset of the 3-K phase for $H = 0$ T and $H \parallel ab$ suggests that a SC state which does not give rise to the ZBCP nucleates at the onset of the 3-K phase.

This interpretation of the absence of the ZBCP near the onset of the 3-K phase for $H = 0$ T and $H \parallel ab$ is supported by the theoretical predictions by Sigrist and Monien.[9] The theory predicts a nucleation of a single-component order parameter with time reversal symmetry conserved. Due to the lower symmetry at the interface, the single-component order parameter with its lobes parallel to the interface k_x, which is illustrated in Fig. 1(a), is favored at the onset of the 3-K phase. Further cooling gives rise to the nucleation of the other component k_y with a relative phase of $\pi/2$, leading to the chiral state $k_x + i\varepsilon k_y$. Because the single-component order parameter k_x does not give rise to the ZBCP, the absence of the ZBCP at the onset of the 3-K phase for $H = 0$ T and $H \parallel ab$ can be attributed to the presence of the single-component order parameter k_x.

The theory also predicts the coupling of the two components of the order parameter in the presence of magnetic field $H \parallel c$. As the two-component state (chiral state) represents a state with a finite orbital angular momentum along the c-axis, a magnetic field component parallel to the c-axis causes the two-component state to be stabilized, unlike $H = 0$ T or $H \parallel ab$. The coupling of the two components is strengthened with decreasing temperature and increasing the magnetic field component parallel to the c-axis. The scenario of the coupling of the two components is supported by recent experimental results.[8] It is possible to consider that the coupling of the two components causes the ZBCP to emerge at the onset of the 3-K phase. The good coincidence of H^* and H_{c2} for $H \parallel c$ can be attributed to the coupling of the two components.

The relation between H^* and H_{c2} shows a distinct difference, depending on the magnetic field direction as discussed above. It is further shown that H^* exhibits in-plane anisotropy when the magnetic field is rotated in the ab-plane. The electric contacts to the individual Ru lamellae enabled the in-plane anisotropy of H^* to be observed. In Fig. 4, the temperature dependence of H^* for the magnetic field parallel and perpendicular to the Ru lamella plane is plotted with open squares and open triangles, respectively. H^* for $H \perp$ Ru-plane is 89% of that for $H \parallel$ Ru-plane at $T = 0.3$ K. On the other hand, no significant dependence of H_{c2} on the field direction relative to the crystallographic axes has been observed in a resistivity measurement in the eutectic system.[21] This is probably because many Ru lamellae oriented in different directions are involved in the resistivity measurement, and thus the anisotropy around each Ru inclusion will be averaged out. Because the 3-K phase is inferred to be surface superconductivity around Ru lamellae,[5,7] the critical magnetic field for $H \perp$ Ru-plane is expected to be smaller than that for $H \parallel$ Ru-plane, as was discussed by Matsumoto *et al.*[20] The observed in-plane anisotropy of H^* is considered to reflect the anisotropy of H_{c2} with respect to the direction of the Ru lamella. The in-plane anisotropy of H^* relative to the Ru lamella directly supports the proposition that 3-K phase superconductivity is filamentary and occurs at surfaces surrounding Ru.[5,7]

To summarize, we have developed a device for the measurement of the conductance at the interface between Sr_2RuO_4 and a single Ru microinclusion. The tunneling spectra with the ZBCP were obtained in the voltage dependence of the differential conductance. The magnetic field and temperature dependence of the onset of the ZBCP is discussed in comparison with the onset of the superconductivity T_c and H_{c2}. We propose that the difference between the onset of the ZBCP and the onset of the 3-K phase can be attributed to the presence of a superconducting state which is not accompanied by the ABS. This is consistent with the theoretical prediction of the nucleation of the single-component state k_x, which does not give rise to the ABS, at the onset of the 3-K phase. We have also observed in-plane anisotropy of H^* with respect to the direction of Ru lamellae.

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